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14. ABSTRACT
This proposal is aimed at investigating the influence of substrate steps on molecular beam epitaxial growth of Bi2Se3 and Bi2Te3 topological insulators on Si (111). It uses a novel combinatorial approach by creating micro-lens structures on Si(111) substrate. The micro-lens contains a combination of numerous micro-surfaces with different miscut angles and miscut orientation. By using scanning probe microscopy, we can investigate how different miscut angles and orientations influence the growth process.

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:

a. REPORT	b. ABSTRACT	c. THIS PAGE	17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
UU	UU	UU	UU		Chih-Kang Shih
					19b. TELEPHONE NUMBER: 512-471-6603

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Prescribed by ANSI Std. Z39.18

Report Title

Final report: Influence of Surface Steps on Molecular Beam Epitaxy of Topological Insulators

ABSTRACT

This proposal is aimed at investigating the influence of substrate steps on molecular beam epitaxial growth of Bi2Se3 and Bi2Te3 topological insulators on Si (111). It uses a novel combinatorial approach by creating micro-lens structures on Si(111) substrate. The micro-lens contains a combination of numerous micro-surfaces with different miscut angles and miscut orientation. By using scanning probe microscopy, we can investigate how different miscut angles and orientations influence the growth process.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
11/29/2013	1.00	Chris Mann, Damien West, Ireneusz Miotkowski, Yong P. Chen, Shengbai Zhang, Chih-Kang Shih. Mapping the 3D surface potential in Bi2Se3, Nature Communications, (08 2013): 0. doi: 10.1038/ncomms3277

TOTAL: **1**

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Number of Presentations: 0.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received		Paper
11/29/2013	2.00	Chris Mann, Damien West, Ireneusz Miotkowski, Yong P. Chen, Shengbai Zhang, Chih-Kang Shih. Observation of Coulomb repulsion between Cu intercalants in CuxBi2Se3, Physical Review Letters (submitted) (10 2013)

TOTAL: **1**

Number of Manuscripts:

Books

Received Paper

TOTAL:

Patents Submitted

One disclosure is submitted at the University of Texas with a tech ID 6407 SHI. Title "A combinatorial substrate for research epitaxy"

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Chris Mann	0.75	
FTE Equivalent:	**0.75**	
Total Number:	**1**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Chris Mann

Total Number: 1

Names of other research staff

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

During the course of the proposal, we have published one paper in Nature Communication and submitted one paper to Physical Review Letters. In addition, one patent disclosure was submitted.

Technology Transfer

Technical Report

Proposal Number: 62395PHII

Proposal Title: Influence of Surface Steps on Molecular Beam Epitaxy of Topological Insulators

Project overview

The modern semiconductor and electronics industries often rely on the use of epitaxial substrates for the fabrication of advanced devices. The deposition and growth of materials can be strongly influenced by a variety of factors including not only the substrate material, but also its crystallographic orientation, substrate miscut angle, temperature, deposition rate, doping profiles, and atomic step morphology; unintentional variation of any of these parameters between different growths can lead to misleading results and greatly impeded rates of research. Furthermore, these substrates are often expensive, making systematic studies very costly.

To reduce the influence of variation between different tests and reduce the number of substrates required to perform a growth study, the proposal is aimed at developing a combinatorial substrate to simultaneously provide multiple atomic step morphologies, miscut angles, and, occasionally, multiple crystallographic orientations, all on a single substrate. In addition we will use this combinatorial substrate to investigate the influence of surface steps on molecular beam epitaxy of topological insulators such as Bi_2Se_3.

Research Accomplishments under the support of the STIR grant

I. Creation of combinatorial substrates

The combinatorial substrates are created out of 4.5-inch Silicon (111) wafers using standard microfabrication techniques (also shown in Fig. 1):

1. A Silicon wafer is coated with photolithographic resist.
2. The wafer is exposed to UV light under a mask with the desired pattern.
3. The exposed photo-resist is removed by acetone and isopropanal alcohol
4. Reflow acetone vapor to the wafer to prepare a smooth texture. This is a known process and can be used to prepare microlenses, for instance[reference].
5. The reflowed resist is then used as an etch mask for plasma etching. The shape of the reflowed resist is transferred into the substrate.
6. The substrate is cleaned of residual resist and is prepared for deposition as though it were a traditional substrate.

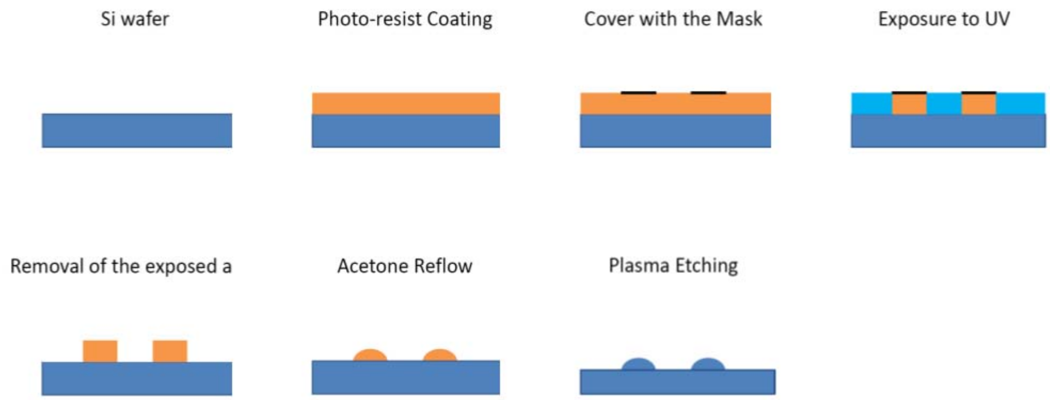

Fig. 1. The schematic showing the procedure to create the microlens array on Si substrate.

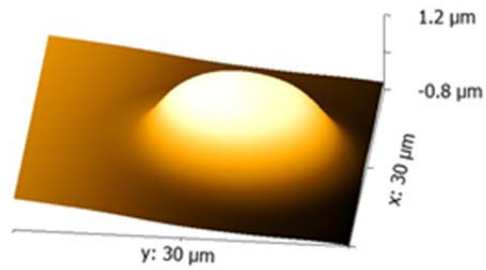

Fig. 2 . An example of an as-processed microlens investigated by AFM.

Right after manufacturing of these micron lenses on silicon, the sample is cleaned using the RCA cleaning process, yielding micron lenses of various whose diameters are defined by the lithography and the heights are defined by the reflow and etching process. Shown in Fig. 2 is an example of a microlens with 20 micron diameter and 0.4 micron height.

The substrate is then transferred into a molecular beam epitaxy UHV chamber with a base pressure < 1 x 10^{-10} torr. The sample is cleaned in-situ by flashing the sample several time up to 1200 C (each time about 10s) to desorb native oxide and other contaminations. The cleanliness of the sample is reflected by the observation of sharp RHEED pattern as shown in Fig. 3. Bi_2Se_3 layers are then grown on such a combinatorial substrate.

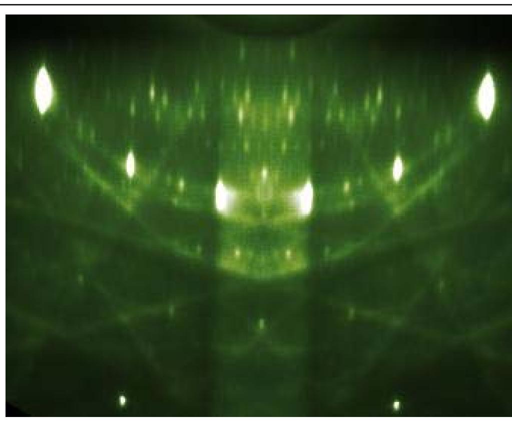

Fig. 3. RHEED pattern of the sample after flashing revealing atomic clean surface.

II. AFM studies of the combinatorial substrate

Atomic force microscopy was used to carry out most of the investigations on the step structures of the combinatorial substrate. Scanning tunneling microscopy (STM) was also used to study the structure in-situ. Nevertheless the field of view of STM is more limited. Most discussions presented here are based on AFM studies.

Fig. 4 shows the AFM image acquired in the region far away from the microlens dome. This region reflects the structure of the original substrate which has a miscut toward ⟨11$\bar{2}$⟩ direction, which is reflected in the regular step structures seen in the image. The average step size is about 30 nm with an average step height of ~ 2nm, corresponding to a bunch of 6 atomic steps (i.e. step bunching). Nevertheless, it also noted that there is also a wide variation of step height distribution.

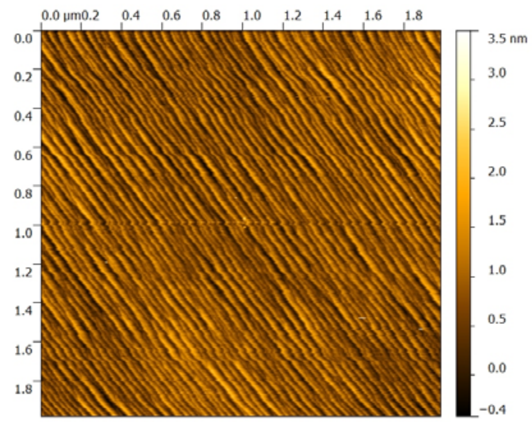

Fig. 4 AFM image of the sample in the substrate region, revealing 4 degree miscut toward ⟨11$\bar{2}$⟩.

Shown in Fig. 5(a) is the AFM image of a microlens dome of ~ 20 µm in diameter and ~ 1.5 µm in height. The highest part of the dome is off-centered along the diameter in ⟨11$\bar{2}$⟩ direction due to the four-degree miscut, and a fairly large one-atomic-step-high terrace forms there. In Fig 5(b) is the zoom-in image around the dome peak. From this zoom in view one can also observe finer steps (most of them near the flat-top region are single atomic steps). We found that for microlens with diameter larger than 20 microns, the dome top deviates from spherical shape, resulting large variation of the shape and location of the flat-top region. On the other hand for those microlenses with diameter smaller than 15 microns, most of them have nice spherical shaped dome, as shown in Fig 5(c).

The symmetrical top terrace also leads to symmetrical distribution of step structures. In this case, a 3-fold symmetry is observed, reflecting the underlying crystalline symmetry. More

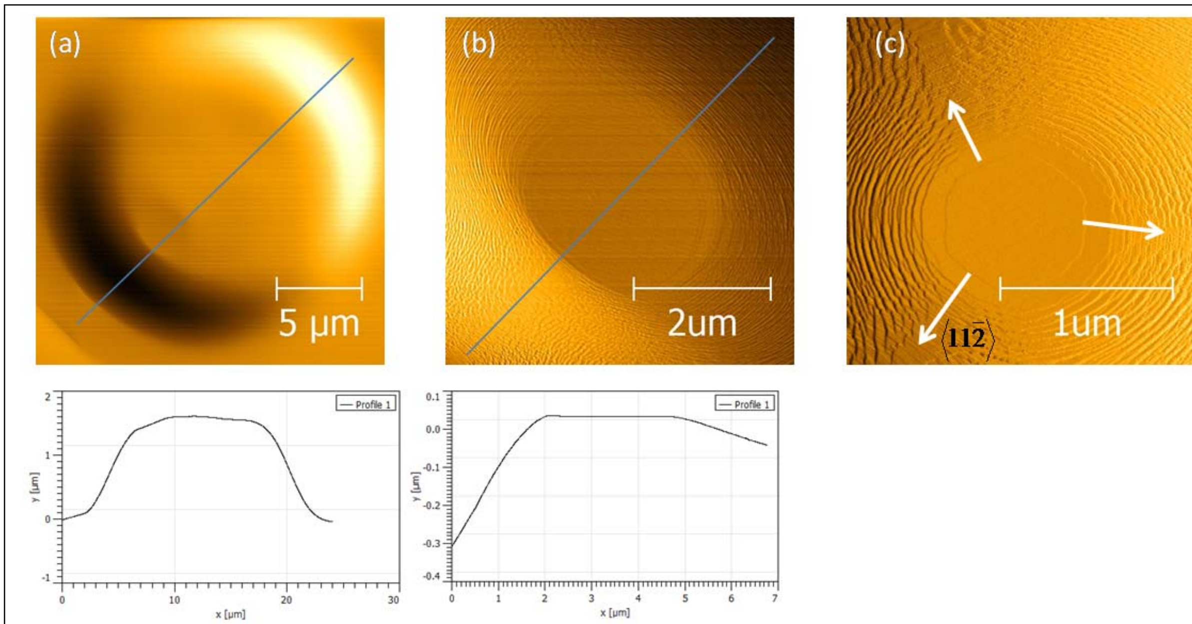

Fig. 5. (a) AFM image (with light shading to enhance the contrast) of a microlens with the lne profile shown below; (b) Zoom in view near the flat top region; (c) AFM image of another microlens with more spherical dome.

interestingly, along three major $\langle 11\bar{2} \rangle$ directions (marked by the three arrows), the steps have straight edge and they are single atomic steps. The straight edge can only sustain certain length and eventually bend over in order to comply with the overall dome shape. Also quite interesting is that along other directions, step bunching phenomena dominate.

III. MBE growth of Bi2Se3 on combinatorial substrate.

Thin Bi2Se3 films are epitaxially grown on the combinatorial substrates in the TI-MBE system. Pure Bi source (99.999%) is evaporated in a commercial Knudsen cell, and pure Se source(99.999%) is evaporated from a home-built evaporator. Bi: Se ratio is kept at 1:10, and the growth rate is around 4 min/QL. The substrate temperature is kept at around 300 ^{0}C. The growth rate is monitored using RHEED oscillations.

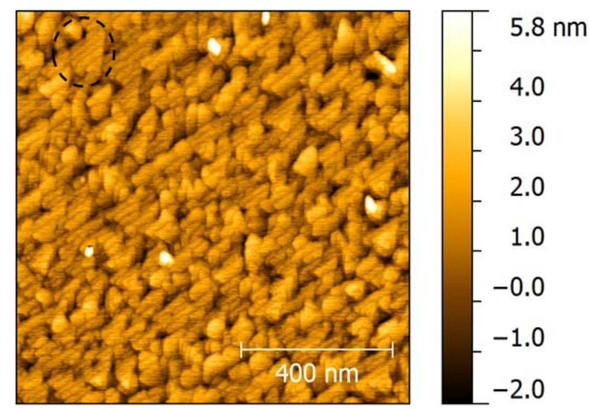

Fig. 6. AFM image of a 8 QL Bi2Se3 film acquired in the substrate region.

After a growth of 8 QL, the samples are investigated using AFM. Shown in Fig. 6 is the image acquired at the “flat” substrate region (with 4 degree miscut). In this region, the film is rough in general, although the surface is interwoven with local regions (one of which is labeled with a dashed circle) that are flat and with well-defined single QL steps with a step height of 1 nm and a step size of ~ 18 nm. We interpreted that these are local regions with the step-bunching height of the substrate to be commensurate with the step-flow growth of QL (we tentatively term it as the “coincident step match”, in analogous to coincident lattice match).

Figure 7(a) shows the AFM image acquired on top of the dome in one of the microlenses. The most obvious feature is the nearly atomically smooth surface in the flat-top region with

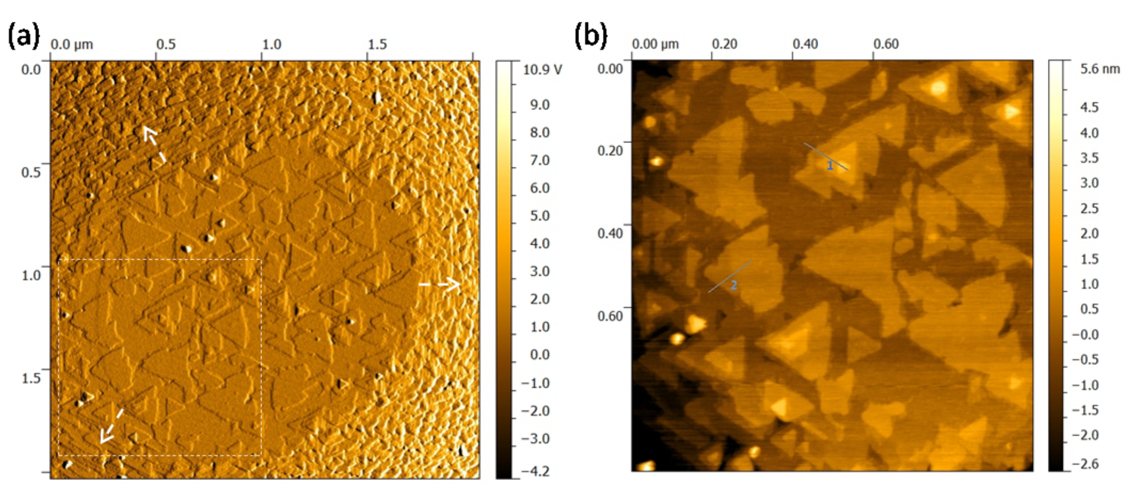

Fig. 7. (a) AFM image of a 8QL Bi2Se3 film grown in the microlens region; (b) a zoom-in view image corresponding to the dashed box in (a).

triangular terraces that are mostly single QL high.

Shown in Fig. 7(b) is the zoom-in view of the region outlined by the dashed box in 7(a). Some of the triangular terraces start nucleating additional layers. The most intriguing aspect is that the newly nucleated terraces appear to start with a reverse triangular shape. This might tell us something about the nature of the nucleation. Right at the edge of the center circular ring, the substrate has a single atomic step. One can observe that the growth of the Bi2Se3 conform to the edge (labeled by line 2). In Fig 7(a), we also label the three <112> directions with dashed arrows. These are regions with straight step edges in the substrate with single step. In these regions, the growth of Bi2Se3 appears to follow a step-flow growth behavior. In other regions, however, one observes primarily rough surface. Note that on microlens surface, the effective miscut angle changes quite rapidly. This leaves little chance for fulfilling the the co-incident step height matching condition, unlike the case for the 4 degree miscut substrate. This makes it more desirable to create micolens substrate with a larger effective radius of curvature so the miscut angle changes more gradually.

IV. Summary

In summary, supported by the ARO STIR grant, we have successfully developed a combinatorial substrate to simultaneously provide multiple atomic step morphologies, miscut angles, and, occasionally, multiple crystallographic orientations, all on a single substrate. The development of this substrate has led to one patent disclosure. In addition, we have carried out MBE growth of Bi2Se3 by using this combinatorial substrate. The result shows new insights into the influence of steps and the miscut direction on the quality of the growth film.